



Missouri University of Science and Technology
Scholars' Mine

Physics Faculty Research & Creative Works

Physics

01 Jan 2005

Microwave Permeability of CO₂ Z Composites

Konstantin Rozanov

Missouri University of Science and Technology, rozanovk@mst.edu

Marina Koledintseva

Missouri University of Science and Technology

Z. W. Li

L. F. Chen

Follow this and additional works at: https://scholarsmine.mst.edu/phys_facwork



Part of the [Electrical and Computer Engineering Commons](#)

Recommended Citation

K. Rozanov et al., "Microwave Permeability of CO₂ Z Composites," *Journal of Applied Physics*, American Institute of Physics (AIP), Jan 2005.

The definitive version is available at <https://doi.org/10.1063/1.1827911>

This Article - Journal is brought to you for free and open access by Scholars' Mine. It has been accepted for inclusion in Physics Faculty Research & Creative Works by an authorized administrator of Scholars' Mine. This work is protected by U. S. Copyright Law. Unauthorized use including reproduction for redistribution requires the permission of the copyright holder. For more information, please contact scholarsmine@mst.edu.

Microwave permeability of Co₂Z composites

K. N. Rozanov^{a)}

Institute for Theoretical and Applied Electromagnetics, Moscow, Russia

Z. W. Li and L. F. Chen

Temasek Laboratories, National University of Singapore, Singapore

M. Y. Koledintseva

Electrical and Computer Engineering Department, University of Missouri–Rolla, Rolla, Missouri

(Received 10 March 2004; accepted 12 October 2004; published online 16 December 2004)

The microwave permittivity and permeability of Co₂Z barium ferrite composite samples are measured as functions of frequency and volume fraction of the ferrite. Magnetostatic properties of the bulk ferrite are determined. This allows Snoek's law [J. L. Snoek, *Physica* **14**, 204 (1948)] to be verified by comparing the microwave and magnetostatic Snoek's constants. The modification of Snoek's law for hexagonal ferrites suggested recently by Acher *et al.* [*Phys. Rev. B* **62**, 11324 (2000)] is also verified. Acher's constant is found from microwave measurements to agree with the value calculated from the magnetostatic properties of bulk ferrite, but microwave and magnetostatic Snoek's constant do not agree. This may be attributed to the effect due to demagnetizing factors of ferrite inclusions that are not considered in the derivation of Snoek's and Acher's laws. The measured frequency-dependent permeability of composites satisfies the Lorentzian dispersion law and is consistent with the Maxwell Garnett approximation [J. C. Maxwell Garnett, *Philos. Trans. R. Soc. London* **203**, 385 (1904)]. According to the theoretical analysis based on the Lorentzian dispersion law and the Maxwell Garnett mixing rule, both Snoek's and Acher's constants must be linear functions of the volume fraction, independent of whether microwave values of the constants are in agreement with the magnetostatic values. In contrast, the experimental measurements reveal a steady decrease of both constants with the volume fraction. The disagreement is discussed in terms of the influence of effective medium in composite on the inherent permeability of ferrite particles.

© 2005 American Institute of Physics. [DOI: 10.1063/1.1827911]

I. INTRODUCTION

Specially designed ferrite particles are one of the best fillers for the development of electromagnetic wave attenuation materials. Ferrite composites are known to provide high level of magnetic loss, high resistivity, and the capability of withstanding intense electromagnetic fields. At the same time, they possess high chemical stability, and their density is low in comparison with that of ferromagnetic metals. Compared to spinel ferrites with a cubic crystallographic structure, hexagonal ferrites, such as Co₂Z, have a higher resonance frequency and a higher microwave permeability, and therefore are more useful in the gigahertz frequency range.^{1,2} Microwave magnetic properties have recently been extensively studied for hexagonal ferrite composites, see Refs. 3–7 and references therein.

In ferrites, the frequency dispersion of permeability is determined by the Landau–Lifshitz–Gilbert equation

$$\frac{\partial \mathbf{M}}{\partial t} = -\mathbf{M} \times \left(\gamma \mathbf{H} - \frac{\alpha}{M_S} \times \frac{\partial \mathbf{M}}{\partial t} \right). \quad (1)$$

In (1), α is the Gilbert damping parameter, M_S is the saturation magnetization, $\gamma=2.8$ GHz/kOe is the gyromagnetic factor, and \mathbf{H} is the total magnetic field, to which the mag-

netic moment \mathbf{M} is exposed. In the absence of magnetic bias, \mathbf{H} includes the demagnetizing fields, the fields associated with magnetic anisotropy, as well as the microwave magnetic field.

For a single-domain ellipsoidal particle with uniaxial anisotropy, Eq. (1) results in a frequency-dependent permeability, $\mu = \mu' - i\mu''$, given by

$$\mu(f) = 1 + \frac{\chi + i\beta f}{1 + if/f_d - (f/f_r)^2}, \quad (2)$$

where f is the frequency and parameters χ , f_d , f_r , and β are functions of the shape and magnetic structure of the particle. The value of $\chi = \mu_{st} - 1$, where μ_{st} is the static permeability, corresponds to the static susceptibility of the magnet. Parameters f_d and f_r are Debye and resonance characteristic frequencies, respectively, that determine the location of the magnetic loss peak and the shape of the magnetic dispersion curve. The values of β and f_d are functions of the Gilbert damping parameter α . Since Eq. (1) is valid at $\alpha \ll 1$, the second term in the numerator of (2) has a negligible effect on the shape of dispersion curve in the vicinity of the resonance. For this reason, this term is usually neglected resulting in the Lorentzian dispersion law

$$\mu(f) = 1 + \frac{\chi}{1 + if/f_d - (f/f_r)^2}. \quad (3)$$

^{a)}Author to whom correspondence should be addressed; electronic mail: k_rozanov@mail.ru

The resonance frequency of a crystallite depends on its shape and orientation. Hence there is a distribution of resonance frequencies in a polycrystalline sample that is dependent on the composition and technology of the material. In most actual magnets, the resultant magnetic dispersion curve is still of the Lorentzian shape but the width of the spectrum increases greatly, being a function of inhomogeneity of the sample rather than being a function of the Gilbert damping parameter. If the spectral spread is due to inhomogeneity, then the location of the magnetic loss peak is determined by the mean resonance frequency. For this case, it is readily shown that f_r in (3) is located near the high-frequency boundary of the distribution of resonance frequencies of crystallites. If the distribution is wide, then $f_d \ll f_r$ and the dispersion law is close to the Debye dispersion law, which can be obtained from (3) at $f_r = \infty$. In Debye dispersion curves, the magnetic loss peak is located at f_d . In the case of $f_d \ll f_r$, the Lorentzian dispersion curve (3) differs from the Debye one only at frequencies near f_r , i.e., much higher than the magnetic loss peak. Following Ref. 8, the Lorentzian dispersion curves with $f_d < f_r$ are referred to in this paper as wideband Lorentzian curves.

If there are several different physical mechanisms contributing to the permeability dispersion in a magnet, then the magnetic spectrum may contain several loss peaks. An example is a combination of the ferromagnetic resonance and the effects arising due to the domain-wall motion. In this case, the magnetic dispersion curve is frequently a sum of two resonance terms:^{9,10}

$$\mu(f) = 1 + \frac{\chi_1}{1 + i f/f_{d1} - (f/f_{r1})^2} + \frac{\chi_2}{1 + i f/f_{d2} - (f/f_{r2})^2}. \quad (4)$$

In ferrites, the term in (4) responsible for the domain-wall motion is typically of the Debye type. Fitting dispersion curves with a sum of several resonance terms is also appropriate if a wide spectrum is distorted significantly compared to the pure resonance law (3), as is frequently observed in actual magnets.⁵ With a properly chosen sum of several resonances, any actual dispersion dependence can be fitted.

To calculate the permeability of ferrite composites, various mixing rules are employed. The simplest and the most well known of these are the Maxwell Garnett approximation¹¹ (MGA) and the effective medium approximation (EMA).¹² For a two-phase mixture comprising spherical ferrite inclusions and a nonmagnetic host matrix, the MGA equation for the effective permeability of the composite μ is

$$\frac{\mu - 1}{1 + (\mu - 1)/3} = p \frac{\mu_f - 1}{1 + (\mu_f - 1)/3}, \quad (5)$$

and the EMA equation is

$$p \frac{\mu_f - \mu}{\mu + (\mu_f - \mu)/3} + (1 - p) \frac{1 - \mu}{\mu + (1 - \mu)/3} = 0, \quad (6)$$

where μ_f is the permeability of bulk ferrite and p is the volume fraction of inclusions. It is assumed in both (5) and (6) that the magnetic properties of ferrite particles are independent of the surroundings.

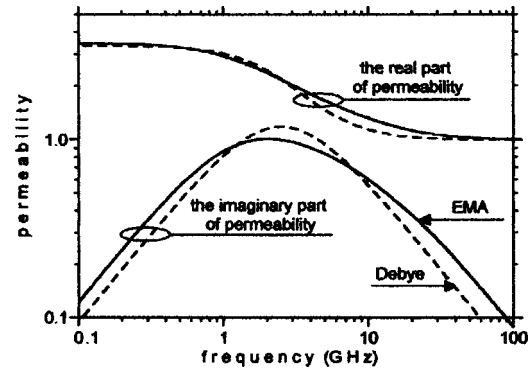


FIG. 1. Frequency dependence of effective permeability calculated with the EMA (6) (solid line). Inclusions in composite are assumed to exhibit the Debye frequency dispersion with $(\chi)_f = 30$, $(f_d)_f = 1$ GHz, and the volume fraction of inclusions is $p = 0.3$. The dotted line is the best fit of the solid line with the Debye dispersion law.

The permeability and permittivity of composite are governed by the same mixing rule. For the case of permittivity, μ_f and μ in (5) and (6) are replaced by the permittivity of the bulk ferrite and the permittivity of the composite, respectively, both are normalized to the permittivity of the host matrix.

If the MGA mixing rule holds for a composite, and if the magnetic dispersion of inclusions is of the Lorentzian type (3), then the dispersion law for the composite will be Lorentzian as well. It can be shown by substituting (3) to (5) that the parameters χ , f_d , and f_r of the dispersion law for the composite are related to those of inclusions, $(\chi)_f$, $(f_d)_f$, and $(f_r)_f$, through

$$\chi = \frac{\chi_f p}{\chi_f (1 - p)/3 + 1}, \quad (7)$$

$$f_d = (f_d)_f [\chi_f (1 - p)/3 + 1], \quad (8)$$

$$f_r = (f_r)_f \sqrt{\chi_f (1 - p)/3 + 1}. \quad (9)$$

If any mixing rule other than MGA is applied to a composite comprising inclusions with the permeability dispersion of the Lorentzian type (or of the Debye type, as a particular case), the resultant dispersion curve of a composite is distorted. As an example, the solid lines in Fig. 1 shows the permeability dispersion curve of a composite obtained by the EMA mixing rule (6). The dispersion curve is calculated under the assumption that inclusions exhibit the Debye frequency dispersion with $\chi_f = 30$ and $(f_d)_f = 1$ GHz, and the volume fraction of the inclusions is $p = 0.3$. The resulting dispersion curve of the composite is fitted with the Debye dispersion law as indicated by the dotted lines. The difference between the solid and dotted lines is evident.

In many bulk magnets, the parameters of the dispersion law relate to each other by Snoek's law¹³

$$(\mu_{st} - 1)f_r = \frac{2}{3} \gamma^4 \pi M_s. \quad (10)$$

The right part of (10) is a definition of Snoek's constant S . By means of (10), Snoek's constant for a bulk magnet can be obtained from the measured dispersion curve. Snoek's law

(10) is valid for magnetic spectra appearing due to both the domain-wall motion and the ferromagnetic resonance. This law is usually suitable for spinel ferrites with a cubic structure and for barium ferrites with an easy c axis of anisotropy. For ferromagnetic resonance, Snoek's law can be derived from (1) provided that the magnetic particle under consideration has a spherical shape and possesses uniaxial anisotropy.

For hexagonal ferrites that exhibit an anisotropy with an easy c plane Snoek's law has a different form,¹⁴

$$(\mu_{st} - 1)f_{\max} = \frac{1}{2} \gamma \times 4\pi M_S \sqrt{H_\theta/H_\phi}, \quad (11)$$

where H_θ and H_ϕ are out-of-plane and in-plane anisotropy fields, respectively.

Recently, Adenot *et al.*¹ suggested another form of Snoek's law for hexagonal ferrites

$$\int_0^\infty f \mu''(f) df = \frac{\pi}{2} \frac{1}{3} (\gamma 4\pi M_S)^2 \left(1 + \frac{H_\theta + H_\phi}{4\pi M_S} \right). \quad (12)$$

Equation (12) is derived from the Landau–Lifshitz equation assuming a Bloch–Blombergen dissipation term, which results in the Lorentzian dispersion law for the permeability. If the permeability is obtained from the Landau–Lifshitz–Gilbert equation as in (2), the integral on the left side of (12) diverges. Thus, (12) is valid for any magnetic dispersion dependence that can be expressed as a sum of Lorentzian dispersion curves with finite resonance frequencies.

If the dispersion curve is a single Lorentzian line, then the integral in the left part of (12) is readily expressed by the parameters of the line, $\chi = \mu_{st} - 1$ and f_r . For composite magnets, it is rigorously proven¹⁵ that the left part of (12) is a linear function of the volume fraction. This is independent of the type of a mixing rule that governs the effective permeability of the composite. Therefore, p must be introduced in the right part of (12) as a factor when a composite is under study. Also, in the cases of practical interest, microwave hexagonal ferrites have $H_\theta \gg H_\phi$, and H_ϕ can be neglected in (12). Then, (12) can be rewritten as

$$(\mu_{st} - 1)f_r^2 = \frac{p}{3} (\gamma 4\pi M_S)^2 \left(1 + \frac{H_\theta}{4\pi M_S} \right). \quad (13)$$

In this paper, (13) is referred to as Acher's law for hexagonal ferrites, by the analogy to Snoek's law. The right part of (13) is Acher's constant, A .

Snoek's and Acher's laws establish the relation between the parameters of magnetic dispersion curves and magneto-static parameters of the magnet. These laws are helpful in the analysis of experimental data. Another reason for the interest in Snoek's and Acher's laws is that they allow constraints on microwave permeability values to be established. Larger Snoek's and Acher's constants result in larger real and imaginary parts of permeability that can be obtained at higher frequencies. Therefore, these constants are a good indicator of microwave performance of magnets.¹⁶

Acher's law in the form (13) is more convenient than the integral form (12) for the analysis of experimental data, since measurements are conducted within a limited frequency band. The contribution of high-frequency permeability val-

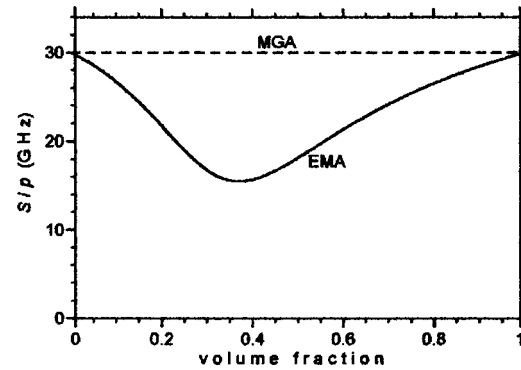


FIG. 2. The ratio of Snoek's constant to the volume fraction, S/p , yielded by EMA (6) with $(\chi)_f = 30$, $(f_d)_f = 1$ GHz, as a function of volume fraction.

ues into the integral may be significant because frequency is involved as a factor in the integrand. Therefore, the integral form (12) can be useful in establishing the constraints on the microwave permeability of magnets. Equation (13) can be applied to establish the relationship between the magneto-static properties of magnets and the parameters of microwave frequency dispersion. It is valid only for the case of magnetic spectra consisting of a single Lorentzian line, which occurs frequently, and, therefore, can be of importance for many practical problems.

Snoek's constant in composites is typically not a linear function of volume fraction. It is readily shown that S will be proportional to p , if the inclusions only exhibit the Debye frequency dispersion and the composite is governed by the MGA mixing rule. As is shown above, in the case of the Debye dispersion, the resonance frequency f_r in (10) must be replaced with the frequency f_{\max} , at which magnetic loss has a peak, and f_{\max} is equal to f_d . After that, the linear dependence of Snoek's constant on p for the MGA mixing rule follows readily from (7)–(9).

According to the above discussion, Snoek's law can be written as

$$(\mu_{st} - 1)f_d = \frac{p}{2} \gamma \times 4\pi M_S \sqrt{H_\theta/H_\phi}, \quad (14)$$

for a composite comprising a hexagonal ferrite and governed by the Debye dispersion law and the MGA mixing rule.

For any other mixing rule as well as for any other dispersion law, Snoek's constant is not proportional to p . As an example, Fig. 2 shows the volume fraction dependence of the ratio S/p as is obtained from the EMA mixing rule (6) with $\chi_f = 30$ and $(f_d)_f = 1$ GHz. In the calculation, f_d in (14) is replaced with f_{\max} . There is a pronounced minimum of the S/p ratio near the percolation threshold, $p_c = 1/3$. The value of S/p obtained with the MGA mixing rule (5) is independent of p , as shown by the dotted line in Fig. 2.

This paper is aimed at the verification of laws (13) and (14) in microwave hexagonal ferrite composites. In Sec. II, techniques for the preparation and microwave measurement of Co₂Z barium ferrite composite samples are described. In Sec. III, measured data on the composites are presented and discussed. It is found that the measured data agree with the Maxwell Garnett approximation and the dispersion curves are of the wideband Lorentzian type. Therefore, both laws

(13) and (14) are expected to be valid for the composites under study. However, some disagreements with the predictions of the theory are observed. In Sec. IV, a possible reason for the disagreement is proposed in terms of the influence of the effective medium in composite on the inherent permeability of an individual ferrite particle.

II. EXPERIMENT

Co_2Z barium ferrite, $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$, is synthesized using conventional ceramic techniques. The size of the ferrite particles is about $10\ \mu\text{m}$. X-ray diffraction has identified that this is a single-phase ferrite with the Z-type hexagonal structure. The lattice parameters are $a=0.588\ \text{nm}$ and $c=5.23\ \text{nm}$.

The sintered sample was prepared in the form of a cylinder with a diameter of $4.0\ \text{mm}$ and the length of about $2\ \text{mm}$. The magnetization curves and M - H loop are measured with a static magnetic bias of 0 – $80\ \text{kOe}$, and between -30 and $+30\ \text{kOe}$, respectively, at room temperature for the sintered sample using the vibrating sample magnetometer made by Oxford Instrument. The demagnetizing correction has been considered. Coercivity H_c was obtained from the M - H loop, and saturation magnetization $4\pi M_s$ is deduced from a numerical analysis of the magnetization curve, based on the law of approach to saturation. The out-of-plane anisotropy field, H_θ , was determined by the initial magnetization curves parallel and perpendicular to the alignment direction for the aligned sample.¹⁷ The field corresponding to the intersection of these two curves is the anisotropy field H_θ . The measurement of magnetostatic properties of the bulk ferrite yields $4\pi M_s=3.3\ \text{kG}$, $H_c=23\ \text{Oe}$, and $H_\theta=12\ \text{kOe}$. No measured data on the in-plane anisotropy field, H_ϕ , are available in this study. It is known from the literature^{14,18} that $H_\phi \approx 0.12\ \text{kOe}$ for Co_2Z barium ferrites.

Co_2Z composites are prepared by mixing the fine powders of Co_2Z ferrite with epoxy resin with a permittivity of 2.9 . The volume fractions of the powders vary from 0.1 to 0.5 . Then, the samples are fabricated into cylinders with the outer diameter of $6.8\ \text{mm}$, inner diameter of $3\ \text{mm}$, and length of about $2\ \text{mm}$ for microwave measurement.

The real and imaginary parts of permeability are measured in the frequency range of 0.5 – $16.5\ \text{GHz}$ using transmission/reflection method.^{19,20} The measurement fixture is a set of $7/3\ \text{mm}$ coaxial air-line with the length of $60\ \text{mm}$. The microwave measurement is conducted with an HP8722D Vector Network Analyzer.

To find the dispersion law from the measured permeability of the composite, fitting of the measured magnetic dispersion curves is made by a numerical minimization of the rms difference between the measured permeability values, μ_{meas} , and the permeability calculated with the chosen dispersion law, μ_{calc} ,

$$\kappa = \frac{1}{N} \sum_{i=1}^N \left\{ \frac{[\mu'_{\text{meas}}(f_i) - \mu'_{\text{calc}}(f_i)]^2}{(\Delta'_i)^2} + \frac{[\mu''_{\text{meas}}(f_i) - \mu''_{\text{calc}}(f_i)]^2}{(\Delta''_i)^2} \right\}, \quad (15)$$

where the summation is over N frequency points in the range

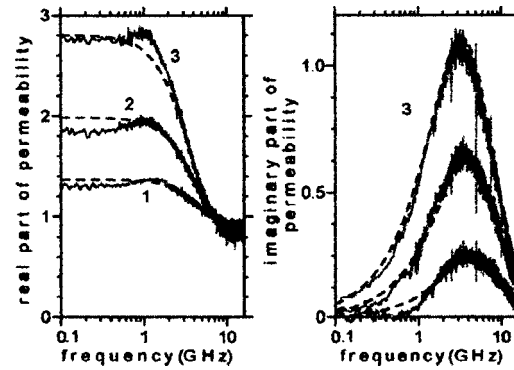


FIG. 3. The measured frequency dependence of permeability (the solid lines, left: the real part, right: the imaginary part) for the Co_2Z composites. The volume fractions of ferrite are 1, $p=0.1$; 2, $p=0.3$; and 3, $p=0.5$. The dotted lines show the results of fitting the measured data with the Lorentzian dispersion law (3).

of measurement. In (15), $(\Delta'_i)^2$ and $(\Delta''_i)^2$ stand for the squared measurement uncertainty of μ' and μ'' , respectively, measured at frequency f_i . The uncertainty is estimated to depend on the measured value through $(\Delta')^2=(0.1)^2+(0.1\mu')^2$ and $(\Delta'')^2=(0.1)^2+(0.1\mu'')^2$.

III. RESULTS AND DISCUSSION

The measured frequency dependence of permeability for three samples under study is shown in Fig. 3. The volume fractions of inclusions for these samples are 0.1 , 0.3 , and 0.5 . It is seen that the permeability exhibits strong frequency dispersion at microwave frequencies. In contrast to the permeability, the permittivity of the samples is almost constant within the measured frequency band, and the dielectric loss is small.

The measured permeability in Fig. 3 is fitted with the Lorentzian dispersion law (3), where the parameters χ , f_d , and f_r are varied until the least value of κ in (15) is achieved. The fitting curves are presented in Fig. 3 by the dotted lines.

Dispersion laws (2) and (4) involve additional fitting parameters as compared to (3). For this reason, the use of these laws as μ_{calc} in (15) yields slightly better agreement with the experiments. However, the volume fraction dependence of the additional fitting parameters is found to be irregular. Therefore, fitting with dispersion laws (2) and (4) does not provide any additional data on the properties of the composites under study.

On the other hand, the resultant value of κ is less than unity for all samples under study for the fitting made with the Lorentzian law. This means that the measurement results are described by the Lorentzian dispersion curves within the measurement accuracy. The Lorentzian dispersion law (3) is therefore supposed to be valid for the composites under study in the discussion below.

Figures 4 and 5 represent the fitted parameters of the Lorentzian dispersion law against the volume fraction p . In Fig. 4, the static permittivity and permeability are given as functions of p . The static permittivity ϵ_{st} is obtained by averaging the measured data over the total frequency range of

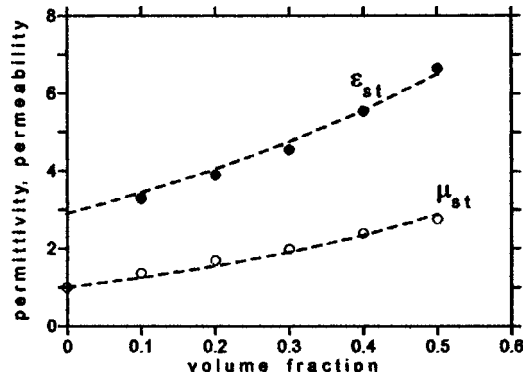


FIG. 4. The static permittivity, ϵ_{st} , and static permeability, μ_{st} , of Co_2Z composites plotted against the volume fraction (dots). The lines are the best fit of the dependences with the MGA equation for spherical inclusions. The material parameters of the inclusions are taken as $\epsilon_i=16$, $\mu_i=11$.

measurement. The static permeability is $\mu_{st}=\chi+1$, where χ is the result of the fitting procedure. In Fig. 5, f_d and f_r are plotted versus p .

For $f_d < f_r$, all the measured dispersion curves are of the wideband Lorentzian type. Therefore, according to the analysis in the Introduction, the MGA mixing rule (5) must govern the properties of the composites. To compare the experimental results with the theory, the material constants of the corresponding bulk material must be known. In this study, such data were unavailable. Herein, the parameters of the Lorentzian dispersion law for the bulk ferrite are varied until the best agreement between the calculated and measured data on the volume fraction dependence of Lorentzian parameters of composites. First, fitting the experimental data for the static permeability of composites with (7) yields the static permeability of bulk ferrite as 11.0. Since the same mixing rule must be valid for the permeability and the permittivity, the same procedure is applied for searching the permittivity. The best agreement yields the value of 15.6 for the permittivity of bulk ferrite. The data obtained are close to published data, see Refs. 21 and 18, respectively.

After that, the characteristic frequencies are found by fitting the experimental data with (8) and (9), provided that the value of $(\chi)_f$ is taken from the best fit of the static permeability. The results obtained are $(f_d)_f=1.0$ GHz and $(f_r)_f$

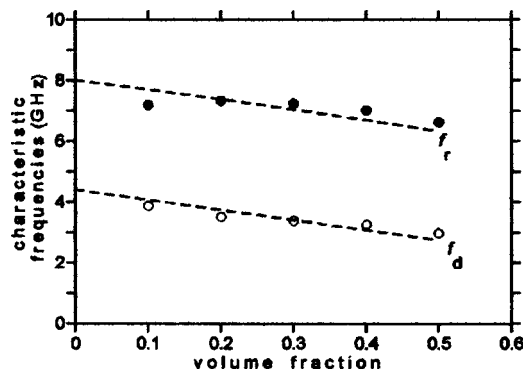


FIG. 5. The Debye and resonance frequencies of Co_2Z composites as functions of the volume fraction (dots). The lines are the best fit of the dependences with the MGA equation for spherical inclusions. The assumptions on the properties of bulk ferrite are $(\chi)_f=10$, $(f_d)_f=1.0$ GHz, and $(f_r)_f=3.9$ GHz.

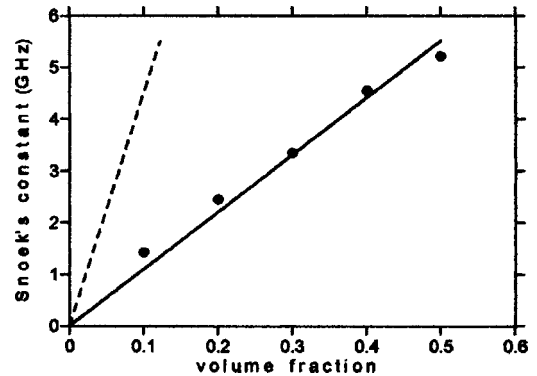


FIG. 6. Measured Snoek's constant S of the composites under study as a function of the volume fraction (dots). The solid line is a linear fit for the measured data. The dotted line is the result of Eq. (9) with $H_\phi=0.12$ kOe.

$=3.9$ GHz. The parameters of the Lorentzian dispersion law obtained by (7)–(9), given the calculated properties of the bulk ferrite, are shown in Figs. 4 and 5 by the dotted lines.

The parameters obtained from the dispersion law allow Snoek's and Acher's constants to be calculated by (14) and (13), respectively. Figure 6 plots the calculated Snoek's constant as a function of volume fraction. The solid line is the best fit to the data with a linear function. By extrapolating this line to $p=1$, Snoek's constant for the bulk ferrite is obtained as 11.1 GHz. If this value of Snoek's constant is taken, then, according to (14), $H_\phi=2.1$ kOe. This disagrees markedly with the reference data,^{14,18} $H_\phi=0.12$ kOe. The values of S calculated using (14) with $H_\phi=0.12$ kOe are shown in Fig. 6 by the dotted line. It is seen that (14) predicts much larger Snoek's constant and therefore much better microwave magnetic performance of the composites under study than is observed in the actual composites. Therefore, it can be deduced that Snoek's law is not valid for composites under study.

This means that the parameters of the measured dispersion curves cannot be related to the magnetostatic properties of the bulk ferrite by means of (14). However, the linear dependence of the left side of (14) on the volume fraction is a consequence of the MGA mixing rule. The validity of the latter is evidenced by the absence of distortions of the measured dispersion curves. Therefore, the left part of (14) must be a linear function of p for the composites under study.

Figure 6 reveals a slight disagreement between the measured Snoek's constant and that obtained under assumption on the linear dependence of S on the volume fraction of inclusions. This disagreement is also clearly demonstrated in Fig. 7, where the ratio of Snoek's constant to the volume fraction, S/p , recalculated from the data in Fig. 6 is plotted against p . The figure reveals a steady decrease of the S/p ratio with p . The extrapolation of these data to $p=1$ produces the value of $S=5.8$ GHz for the bulk ferrite. The use of this value in (14) results in $H_\phi=7.6$ kOe that is in an even greater disagreement with the reference data than the above result. Notice that the extrapolated value of S is in close agreement with $S=6.2$ GHz obtained from ordinary Snoek's law (10).

Notice also that much more realistic value for the in-plane anisotropy field, $H_\phi=0.17$ kOe, follows from the extrapolated value of S with true resonance frequency f_r intro-

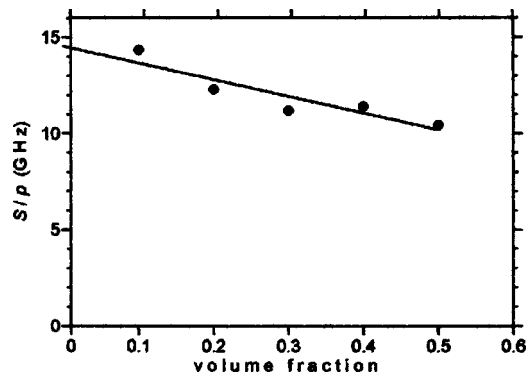


FIG. 7. The measured ratio of Snoek's constant to the volume fraction, S/p , plotted against the volume fraction (dots). The lines show the linear fit of the measured data.

duced in Snoek's law (14) instead of f_d . A possible reason for this can be that the width of the magnetic spectrum is determined mostly by the intrinsic damping parameter of the material. In this case, the contribution to the linewidth from the external damping, i.e., from the spread of resonance frequencies in different areas of a ferrite, is negligible. This could distinguish the ferrite under study from most ferrites, where the external damping is dominant. However, if it is so, then Snoek's constant is not a linear function of volume fraction as it follows from (9), and Snoek's law for composites is not valid anyway.

Figure 8 shows how Acher's law (13) is held in the composites under study. Although the observed dispersion curves are of the wideband Lorentzian type and the primary features of the dispersion curves are due to the Debye frequency, the resonance frequency is determined precisely from the measured data as the frequency where the real part of permeability equals to unity. Shown in the figure are the values of Acher's constant A obtained by substitution of the measured parameters of dispersion curves to the left part of (13) and normalized to the volume fraction of inclusions. As is discussed above, these normalized values must keep constant with the volume fraction for any mixing rule, provided that the Lorentzian law for magnetic dispersion is valid. However, it is seen from the figure that the experimental values of A/p decrease with p .

The extrapolation of the dependence of A as a linear

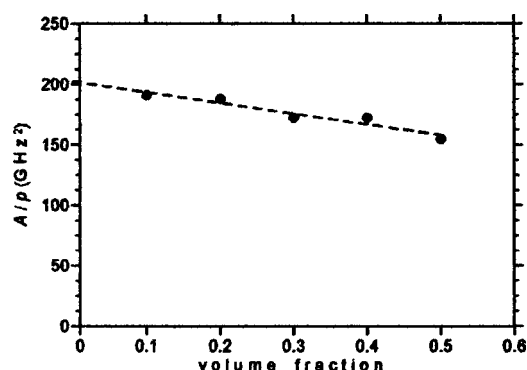


FIG. 8. The measured ratio of Acher's constant to the volume fraction, A/p , plotted against the volume fraction (dots). The lines show the linear fit of the measured data.

function of the volume fraction to $p=1$ yields $A \approx 120 \text{ GHz}^2$ in a good agreement with the value 130 GHz^2 obtained from (13) with $H_\theta=0.12 \text{ kOe}$. This differs from the data from Ref. 1, where the measured values of Acher's constant are between 1/2 and 2/3 of the value yielded by (13). The difference can be attributed to the different approaches to the determination of Acher's constant. In this paper, it is determined from the parameters of the Lorentzian dispersion law by (13). In Ref. 1, Acher's constant is obtained from the integral law (12). The effect of high-frequency permittivity values, which are beyond the frequency band of measurement but can contribute greatly to the integral, may be a reason for obtaining the diminished value of Acher's constant from the measurements in Ref. 1.

IV. CONCLUSION

In this paper, Snoek's and Acher's laws are verified for Co_2Z hexagonal ferrite composites. The values of Snoek's constant as obtained from microwave measurements are smaller than that obtained from magnetostatic properties of a bulk ferrite. Therefore, Snoek's law does not hold for the composites under study. The values of Acher's constant obtained by these approaches are in good agreement. Acher's law is thus found to be valid.

The reason for invalidity of Snoek's law may be that the Landau-Lifshitz equation involve demagnetizing factors of magnetic inclusions. These demagnetizing factors have an effect on the parameters of magnetic dispersion. Therefore, the demagnetizing factors must be involved in the right parts of Snoek's and Acher's laws. The effect of the demagnetization on the law, which is analogous to (12), and is valid for thin magnetic films, is discussed in Ref. 15. Notice that Acher's constant obtained for low concentrations of inclusions is larger than the theoretical value, which is considered as a constraint on the microwave permeability of hexagonal ferrites. Hence, the study of the effect of the demagnetizing factors on Acher's constant may be useful in view of the problem of developing hexagonal ferrites with high microwave permeability.

Another possible reason could be that the intrinsic damping is dominant, as is discussed above. It is a common opinion that intrinsic damping and extrinsic damping are indistinguishable from the experimental data.²² The data obtained reveal that the analysis based on Snoek's law could allow these to be distinguished.

The microwave permeability of the composites under study is in agreement with the predictions of the MGA mixing rule. However, determination of Snoek's and Acher's constants from the measured data reveals disagreement with the theory, which are not seen from comparison of measured and calculated dispersion curves. In the theory, both Snoek's and Acher's constants, normalized to the volume fraction, must be independent of the volume fraction. In actual composites, a steady decrease of both the normalized constants with the volume fraction is observed, which cannot be attributed to the uncertainty of the measurement and to the inaccuracy of the fitting procedure.

This disagreement can also be attributed to the effect of

demagnetizing fields. If a nonspherical magnetic particle is considered as an inclusion in a composite, then the demagnetization fields in this inclusion appear to be dependent on the surrounding.²³ Therefore, the basic assumption on which all the mixing rules are based, namely, the material properties of inclusions are independent on the surroundings, may not be valid for magnetic composites. Therefore, the theory of magnetic composites must take into account that intrinsic permeability of inclusions may change with the increase or decrease of volume fraction due to the change in demagnetizing fields.²³ For large multidomain inclusions, this correction is small, since the domain structure reduces demagnetizing fields of the particle. The same is true in cases when the shape of inclusions is close to spherical. In contrast, for fine powders and especially for nanocomposites embedding nonspherical single-domain inclusions, the correction may drastically change the magnetic performance of the composite.

Several opportunities to obtain more results on the problem are suggested. First, it is of interest to study composites comprising finer ferrite powders, where more pronounced volume fraction dependence of Snoek's and Acher's constants is expected. Second, it would be helpful to treat composites exhibiting the resonance dispersion of permeability with a higher quality factor to provide more accurate values of the measured Acher's constant. Finally, the use of more precise measurement techniques, e.g., the permeability measurement in a shorted coaxial line, would allow possible deviations of measured dispersion curves from the Lorentzian dispersion law to be traced. From the data obtained, useful information could be extracted on dispersion laws and mixing rules in ferrite composites.

ACKNOWLEDGMENTS

One of the authors (K.N.R.) is grateful to the Russian Federation President Foundation for partial support of the

work according to Grant No. 1694.2003.2. The authors thank Dr. Richard DuBroff (University of Missouri–Rolla) for useful discussions and comments.

- ¹A. L. Adenot, O. Acher, T. Taffary, and L. Longuet, *J. Appl. Phys.* **91**, 7601 (2002).
- ²A. A. Kitaytsev and M. Y. Koledintseva, *IEEE Trans. Electromagn. Compat.* **41**, 15 (1999).
- ³P. Singh, V. K. Babbar, A. Razdan, R. K. Puri, and T. C. Goel, *J. Appl. Phys.* **87**, 4362 (2000).
- ⁴S. P. Ruan, B. K. Xu, and H. Suo, *J. Magn. Magn. Mater.* **212**, 175 (2000).
- ⁵M. Wu, H. Zhang, X. Yao, and L. Zhang, *J. Phys. D* **34**, 889 (2001).
- ⁶P. Lubitz and F. Z. Rachford, *J. Appl. Phys.* **91**, 7613 (2002).
- ⁷M. R. Meshram, N. K. Agrawal, B. Sinha, and P. S. Misra, *J. Mater. Sci.* **25**, 169 (2002).
- ⁸M. Y. Koledintseva, K. N. Rozanov, G. Di Fazio, and J. L. Drewniak, in *Proceedings of the Fifth International Symposium on Electromagnetic Compatibility, EMC Europe 2002, Sorrento, Italy, 9–13 September 2002* (IEEE, NY, 2002), pp. 687–692.
- ⁹T. Nakamura and K. Hatakeyama, *IEEE Trans. Magn.* **36**, 3415 (2000).
- ¹⁰T. Nakamura, *J. Appl. Phys.* **88**, 348 (2000).
- ¹¹J. C. Maxwell Garnett, *Philos. Trans. R. Soc. London, Ser. A* **203**, 385 (1904).
- ¹²D. A. G. Bruggeman, *Ann. Phys. (Leipzig)* **24**, 636 (1935).
- ¹³J. L. Snoek, *Physica (Amsterdam)* **14**, 204 (1948).
- ¹⁴J. Verweel, in *Magnetic Properties of Materials*, edited by J. Smit (McGraw Hill, NY, 1971), p. 101.
- ¹⁵O. Acher and A. L. Adenot, *Phys. Rev. B* **62**, 11324 (2000).
- ¹⁶N. A. Buznikov, A. L. Rakhmanov, and K. N. Rozanov, *IEEE Trans. Magn.* **38**, 3123 (2002).
- ¹⁷Z. W. Li, L. F. Chen, and C. K. Ong, *J. Appl. Phys.* **92**, 3902 (2002).
- ¹⁸P. Allegri, D. Autissier, and T. Taffary, *Key Eng. Mater.* **132–136**, 1424 (1997).
- ¹⁹A. M. Nicolson and G. F. Ross, *IEEE Trans. Instrum. Meas.* **19**, 377 (1970).
- ²⁰W. B. Weir, *Proc. IEEE* **62**, 33 (1974).
- ²¹H.-I. Hsiang and H.-H. Duh, *J. Mater. Sci.* **36**, 2082 (2001).
- ²²J. Huijbregtse, F. Roozeboom, J. Sietsma, J. Donkers, T. Kuiper, and E. van de Riet, *J. Appl. Phys.* **83**, 1569 (1998).
- ²³R. Ramprasad, P. Zurcher, M. Petras, M. Miller, and P. Renaud, *Phys. Status Solidi B* **233**, 31 (2002).